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⑯ Formaldehydbindendes Mittel

Es wird ein formaldehydbindendes Mittel für die Verwendung in flächenförmigen Holzcellulosematerialien, wie Spanplatten, Sperrholz oder Tischlerplatten, in denen ein Klebstoff auf Formaldehydbasis verwendet wird, beschrieben. Das formaldehydbindende Mittel enthält eine organische Hydroxylverbindung und ein Amid.

Glycerin
Harnstoff } in wässr. Lsg.

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A formaldehyde binder

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A Formaldehyde Binder

Boards such as particleboard, chipboard and the like are prepared from lignocellulosic materials using adhesives. The preferred adhesives (or glues) are 10 based on formaldehyde, such as urea-formaldehyde, melamine-formaldehyde, phenol-formaldehyde and resorcinol-formaldehyde resins or mixtures thereof. It is a well-known fact that boards prepared using these adhesives have a formaldehyde odour which is both harmful and unpleasant. Formaldehyde is emitted both during the production of such boards and during their storage and final use. 15

Many methods of avoiding the emission of formaldehyde have been proposed, but all of them are either 20 ineffective or reduce the properties of the boards or require complicated application procedures. Some of these procedures involve spraying or spread coating the warm boards coming out of the press with various solutions, such as solutions of urea and/or ammonia or of ammonium 25 salts. Generally, these types of methods are not desirable for industrial application because they require additional process steps and anyway are not very efficient.

Other methods involve the use of very complicated mixtures of a large number of components, some of 30 which are natural glues. These products also are not very efficient. One handicap thereof is the fact that the properties of natural products are not constant.

Another way of reducing the content of free formaldehyde involves the use of an aqueous suspension of urea prills coated with a special wax. This method, too, requires a separate feeding line, because the product is not 5 added to the adhesive formulation itself.

An object of the present invention is to provide a formaldehyde binder which effectively reduces the formaldehyde odour without reducing the properties of the board, without changing the reactivity of the 10 adhesive formulations and without requiring any additional steps in the production of particleboard, plywood or blockboard.

The invention provides a formaldehyde binder when for use in boards prepared from lignocellulosic materials using adhesives based on formaldehyde, which comprises a solution of

(a) at least one organic hydroxy compound with the exception of C_{1-4} monohydric aliphatic alcohols and
(b) at least one amide
20 in water. The formaldehyde binder can also contain (c) an organic compound which acts as a solvent for (a) and (b) and also reacts with formaldehyde and/or (d) an inorganic compound soluble in water. The organic compound (c) is preferably a C_{1-4} monohydric aliphatic alcohol. The inorganic compound (d) is preferably a halide salt. Even if the individual 25 components are not soluble in water, they may dissolve in water if a mixture thereof is heated in water to 70 °C.

Preferably the organic hydroxy compounds [component (a)] are soluble in water or in C_{1-4} monohydric aliphatic alcohols. Examples of such preferred hydroxy compounds are the dihydric, trihydric and pentahydric alcohols containing up to 6 carbon atoms, the monosaccharides containing up to 6 carbon atoms, the di-

saccharides containing up to 12 carbon atoms and polysaccharides having an Ostwald viscosity up to 200 mPas at 25 °C at a concentration corresponding to 37 % (refraction). Other examples of preferred hydroxy compounds are the 5 aromatic alcohols and phenols, which preferably are used alone or in combination with one or more of the above-mentioned dihydric, trihydric and/or pentahydric alcohols and/or monosaccharides, disaccharides and/or polysaccharides. The phenols and aromatic alcohols can be monohydric or 10 polyhydric phenols and monohydric or polyhydric aromatic alcohols containing one benzene ring. [By the term "aromatic alcohols" we mean alcohols (as compared to phenols) which contain an aromatic group].

Specific examples of suitable organic hydroxy 15 compounds are monoethyleneglycol, diethyleneglycol, glycerine, pentaerythritol, fructose, mannose, sorbitol, dextrose, sucrose, maltose, lactose, dextrin, phenol, resorcinol, hydroquinone and the like.

Preferably the amides [component (b)] used in the 20 formaldehyde binder of the invention are likewise soluble in water or in C_{1-4} monohydric aliphatic alcohols. Particularly preferred are the aliphatic amides containing up to 6 carbon atoms and the aromatic amides containing one benzene ring.

25 Suitable examples of amides are urea, thiourea, formamide, acetamide, benzamide, oxamide, succinamide, malonamide and the like.

If desired, to enhance solubility, the formaldehyde binder of the invention can additionally contain additives 30 [component (c)] which are C_{1-4} monohydric aliphatic alcohols such as methanol, ethanol, isopropanol and the like.

A cheaper and more efficient formaldehyde binder

is obtained if water-soluble inorganic compounds [component (d)] which are preferably halide salts, more preferably halides of alkali metals or alkaline earth metals, such as sodium chloride, potassium chloride and calcium chloride, are added.

5 The ratio of organic hydroxy compound [component (a) and component (c), if present] and inorganic compound [component (d), if present] to amide [component (b)] is preferably 10:100 to 400:100, particularly 10:100 to 200:100, by weight. The formaldehyde binder of the invention can be added to the usual glue formulations in quantities varying from 1 to 10 %, preferably 3 to 7 %, of formaldehyde binder solids, based on the weight of the liquid resin containing 65 % by weight of resin solids.

15 The formaldehyde binder of the invention may contain 20 to 80 % by weight, preferably 50 to 70 % by weight, of the active ingredients [components (a) and (b) and components (c) and/or (d), if present]. The water content of the formaldehyde binder depends on the solubility of the active ingredients and the amount of water which can 20 be tolerated in the glue formulations.

25 The formaldehyde binder of the invention can be produced by simply adding the active ingredients and water to a mixer and mixing until the active ingredients are dissolved. This can be done at room temperature or at an elevated temperature up to 70 °C.

30 The formaldehyde binder of the invention may be used whenever boards are prepared from lignocellulosic materials using adhesives based on formaldehyde, such as urea-formaldehyde, melamine-formaldehyde, phenol-formaldehyde or resorcinol-formaldehyde resins or mix-

tures thereof.

When using the formaldehyde binder of the invention, it is possible to produce boards actually containing 5 less than 10 mg of free formaldehyde per 100 g of the dry board, as determined by the F.E.S.Y.P. (Fédération Européenne des Syndicats des Fabricants de Panneaux de Particules) perforator method No. EN 120.

10 The amount of the free formaldehyde reduction depends on many factors and, therefore, can vary widely. If the emission of free formaldehyde is high (higher than 50 mg of formaldehyde per 100 grams of dry board), the reduction can be as high as up to 60 to 85 %. If the emission 15 of free formaldehyde is relatively low, i.e. 20 to 50 mg of free formaldehyde per 100 g of the dry board, the maximum reduction is usually 50 to 60 %. The amount of reduction obtained also depends on the amount of formaldehyde binder used: The more formaldehyde binder is 20 used, the lower is the amount of free formaldehyde.

When the active ingredients of the formaldehyde binder are used in combination with each other, the free formaldehyde reduction is surprisingly much higher than 25 the sum of the effects of the separate components, and they have no adverse effect on the reactivity of the glue formulation or the properties of the boards.

The following examples illustrate the invention.
30 Parts and percents are by weight.

Example 1

In this example the organic compound containing hydroxyl groups is glycerine and the amide is urea. Here, there is illustrated the synergistic behaviour of these two compounds. Various glue formulations are prepared and each is used subsequently in order to produce particleboard.

The control does not include any of the components of the formaldehyde binder according to the invention. Sample 1 includes both glycerine and urea, sample 2 includes only glycerine and sample 3 includes only urea.

It is noticed from the tables reported here below that glycerine when used on its own (sample 2) is a very efficient formaldehyde binder, but urea (sample 3) gives a poorer formaldehyde reduction and lower mechanical values and water resistance, however, when urea is used in combination with glycerine (sample 1), it gives values that are equivalent to those of glycerine on its own.

We may therefore use a cheaper and less efficient product (urea) and make it react as effectively as a more expensive and more efficient product (glycerine). The efficiency refers to formaldehyde absorbing capacities as well as to maintaining good mechanical properties and water resistance without changing the reactivity of the glue formulation and without the need to introduce any special apparatus for its use.

Formaldehyde reduction actually is in this

case 46 %.

The formulations of the various samples used are as follows:

		<u>1</u> parts by <u>weight</u>	<u>2</u> parts by <u>weight</u>	<u>3</u> parts by <u>weight</u>
5	Glycerine (100 %)	270	590	-
10	Urea (100 %)	320	-	590
10	Water	<u>410</u> 1000	<u>410</u> 1000	<u>410</u> 1000
	% solids	59	59	59

The glue formulations used are as follows:

		<u>Control</u> <u>grams</u>	<u>1</u> <u>grams</u>	<u>2</u> <u>grams</u>	<u>3</u> <u>grams</u>
15	Urea-formaldehyde resin 65 %	3077	3077	3077	3077
20	(Molar ratio F:U 1.27:1)				
	Hardener (Ammonium chloride 15 %)	400	400	400	400
25	Paraffin emulsion 50 %	250	250	250	250
	Ammonia 25° Baumé	5	5	5	5
	Sample 1	-	308	-	-
30	Sample 2	-	-	308	-
	Sample 3	-	-	-	308
	Water	<u>268</u>	<u>-</u>	<u>-</u>	<u>-</u>
	Total	4000	4040	4040	4040
	Gel time in secs.	68	68	71	62

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, and 9 and 8 secs./mm. The thickness of the boards is 17.3 mms.

5 The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 cms.

10 The results obtained are reported in the following table and are average values.

	Control	1	2	3
Density (kg/m ³)	683	669	663	657
Bending strength (N/mm ²)	19.9	18.1	17.3	16.3
15 Tensile strength (N/mm ²)	0.73	0.72	0.71	0.63
2 hr. thickness swelling (%)	5.2	4.9	4.5	5.4
24 hr. thickness swelling (%)	51.4	49.9	47.4	53.1
Free formaldehyde (mg/100 g dry board)	15.8	8.5	8.7	11.7

20

Example 2

This is another example illustrating the synergistic behaviour of glycerine and urea in reducing the free formaldehyde of particleboards while maintaining the mechanical properties of the boards and the water resistance as well.

The control does not include any of the components of the formaldehyde binder according to the invention. Sample 1 contains both components of the formaldehyde binder according to the invention and sample 2 contains only one of the two components (the most efficient of the two components).

35 It is noticed here again that only sample 1 gives free formaldehyde that is below 10 mg/100 g. dry board (which is the desir-

ed level for El class) and is the only one to have absolutely equivalent mechanical properties and water resistance. Formaldehyde reduction actually is 34 %.

5 The formulations of the various samples used are as follows:

		<u>1</u> <u>parts by weight</u>	<u>2</u> <u>parts by weight</u>
	Glycerine (100 %)	128	128
10	Urea (100 %)	424	-
	Water	<u>448</u>	<u>872</u>
		1000	1000
	% solids	55.2	12.8

15 The glue formulations used are as follows:

		<u>Control</u> <u>grams</u>	<u>1</u> <u>grams</u>	<u>2</u> <u>grams</u>
	Urea formaldehyde			
20	resin 65 % (Molar ratio F:U = 1.27:1)	3077	3077	3077
	Hardener (Ammonium chloride 25 %)	400	400	400
25	Paraffin emulsion 50 %	250	250	250
	Ammonia 25° Baumé	5	5	5
	Sample 1	-	268	-
30	Sample 2	-	-	268
	Water	268	-	-
	Total	4000	4000	4000
	Gel time in secs.	66	67	69

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and 8 secs./mm. The thickness of the boards is 17.3 mms.

5 The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 cms.

10 The results obtained are reported in the following table and are average values.

	<u>Control</u>	1	2
Density (kg/m ³)	685	684	687
Bending strength (N/mm ²)	20.7	20.6	19.4
15 Tensile strength (N/mm ²)	0.74	0.73	0.67
2 hr. thickness swelling (%)	11.0	7.9	8.6
24 hr. absorption (%)	23.5	23.2	23.7
Free formaldehyde (mg/100 g dry board)	14.3	9.5	12.3

20

Example 3

This example illustrates the efficiency of mono-ethyleneglycol together with urea as a formaldehyde binder.

25

Two formulations are prepared: the control without any of the ingredients of the formaldehyde binder according to the invention and sample 1 which includes both monethylene glycol and urea.

30

Boards are produced from these two glue formulations and it is proved here, too, that with the formaldehyde binder according to our invention we obtain with a urea-formaldehyde resin that gives

normally boards classified as E 2 (control), boards classified as E 1 (sample 1).

The formaldehyde reduction is in this case
5 37 %.

The formulation of sample 1 used is as follows:

	<u>Sample 1</u>
	parts by weight
10	
Monoethyleneglycol 100 %	360
Urea 100 %	365
Water	<u>275</u>
	1000
15 % solids	72.5

The glue formulations used are as follows:

	<u>Control</u>	<u>1</u>
	<u>grams</u>	<u>grams</u>
20		
Urea formaldehyde resin 65 %	3077	3077
(Molar ratio F:U = 1.27:1)		
Hardener	400	400
(Ammonium chloride 15 %)		
25		
Paraffin emulsion 50 %	250	250
Ammonia 25° Baumé	5	5
Sample 1	-	268
Water	268	-
Total	4000	4000
30		
Gel time in secs.	66	60

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and

8 secs./mm. The thickness of the boards is 17.3 mms.

The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 mms.

5

The results obtained are reported in the following table and are average values.

		<u>Control</u>	<u>1</u>
10	Density (kg/m ³)	685	684
	Bending strength (N/mm ²)	20.7	20.2
	Tensile strength (N/mm ²)	0.74	0.74
	2 hr. thickness swelling (%)	11.0	8.6
	24 hr. absorption (%)	23.5	22.6
15	Free formaldehyde (mg/100 g dry board)	14.3	9.0

Example 4

In this example we are illustrating the
20 synergistic behaviour of monoethyleneglycol and urea.

Boards are prepared from three different formulations: the control wherein no ingredients of the formaldehyde binder according to the invention
25 are used, sample 1 wherein both ingredients of the formaldehyde binder of the present invention are used and sample 2 wherein only one ingredient is used.

It is obvious from the results obtained and
30 reported here in below that sample 1 containing both ingredients is much more effective than sample 2 containing only one ingredient (the most efficient of the two ingredients).

The formaldehyde reduction obtained in this case is 32 %.

The formulations of the various samples used 5 are as follows:

		1	2
		<u>parts by weight</u>	<u>parts by weight</u>
	Monoethyleneglycol 100 %	230	580
10	Urea 100 %	350	-
	Water	<u>420</u>	<u>420</u>
		1000	1000
	% solids	58	58

15 The glue formulations are as follows:

		Control	1	2
		<u>grams</u>	<u>grams</u>	<u>grams</u>
	Urea formaldehyde	3077	3077	3077
20	resin 65 %			
	(Molar ratio F:U = 1.27:1)			
	Hardener	400	400	400
	(Ammonium chloride 15 %)			
	Paraffin emulsion 50 %	250	250	250
25	Ammonia 25° Baumé	5	5	5
	Sample 1	-	268	-
	Sample 2	-	-	268
	Water	<u>268</u>	<u>-</u>	<u>-</u>
	Total	4000	4000	4000
30	Gel time in secs.	70	71	76

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and

8 secs./mm. The thickness of the boards is 17.3 mms. The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 mms.

5

The results obtained are reported in the following table and are average values.

	Control	1	2
10 Density (kg/m ³)	688	688	687
Bending strength (N/mm ²)	17.6	17.6	17.5
Tensile strength (N/mm ²)	0.55	0.60	0.56
2 hr. thickness swelling (%)	6.7	4.9	4.7
24 hr. absorption (%)	20.1	20.1	19.5
15 Free formaldehyde (mg/100 g dry board)	15.0	10.3	12.3

Example 5

In this example the use of a resin of different molar ratio is illustrated as well as various levels of addition of the formaldehyde binder itself.

The formaldehyde binder used has the following formulation:

25

	<u>parts by weight</u>
Glycerine 100 %	270
Urea 100 %	318
Water	<u>412</u>
30	1000
% solids	58.8

The glue formulations used in the various samples are as follows:

		Control	1	2	3
		grams	grams	grams	grams
	Urea formaldehyde resin 65 %	3077	3077	3077	3077
	(Molar ratio F:U = 1.4:1)				
5	Hardener	293	380	380	380
	(Ammonium chloride 15 %)				
	Paraffin emulsion 50 %	250	250	250	250
	Ammonia 25° Baumé	5	5	5	5
	Formaldehyde binder	-	154	215	375
10	Water	375	134	73	-
	Total	4000	4000	4000	4087
	Gel time in secs.	73	73	72	75

Single-layer boards are prepared in the
15 laboratory by spraying each of these formulations onto
25 kgs wood chips. Boards are pressed at 10, 9 and
8 secs./mm. The thickness of the boards is 17.3 mms.
The temperature of the press is 200 °C and the pressure
is 35 kg/cm². The dimensions of the boards produced
20 are 40 x 56 cms.

The results obtained are reported in the
following table and are average values.

		Control	1	2	3
25	Density (kg/m ³)	680	687	685	688
	Bending strength (N/mm ²)	22.0	22.5	22.3	21.6
	Tensile strength (N/mm ²)	0.61	0.63	0.66	0.64
	2 hr. thickness swelling (%)	10.1	9.3	8.8	9.5
30	24 hr. thickness swelling (%)	20.6	21.8	21.0	21.5
	Free formaldehyde (mg/100 g dry board)	22.2	13.0	10.1	9.5

It is noticed that the mechanical properties

and water resistance of the boards are equivalent and the formaldehyde reduction is 41 % in case of sample 1, 55 % in case of sample 2 and 57 % in case of sample 3.

5

Example 6

In this example six different types of poly-alcohols are illustrated, two different types of amides, one additive and a variation in the ratio of alcohol to amide covering a range from 57.5/100 to 385/100.

10

The various types of formaldehyde binder used are as follows: *See Table*

	1	2	3	4	5	6
Dextrose	230	-	-	-	-	-
Diethyleneglycol	-	330	-	-	-	-
Monoethyleneglycol	-	-	260	-	-	110
Glycerine	-	-	-	500	-	-
Sucrose	-	-	-	-	-	110
Sorbitol	-	-	-	-	140	-
Methanol	-	-	200	-	140	80
Urea	400	300	-	130	350	330
Thiourea	-	-	170	-	-	-
Water	<u>370</u>	<u>370</u>	<u>370</u>	<u>370</u>	<u>370</u>	<u>370</u>
Total	1000	1000	1000	1000	1000	1000
% solids	63	63	63	63	63	63
Weight ratio of alcohol/amide	57.5/100	110/100	270/100	385/100	80/100	91/100

All above-mentioned figures are in parts by weight.

The glue formulations used in the various 5 samples are as follows:

	Control	1	2	3	4	5	6
	grams	grams	grams	grams	grams	grams	grams
Urea-formaldehyde resin 65 % (Molar ratio F:U = 1.27:1)	3077	3077	3077	3077	3077	3077	3077
Hardener (Ammonium chloride 15 % solution)	400	500	500	500	500	500	500
Paraffin emulsion 50 %	250	250	250	250	250	250	250
Ammonia 25° Baumé	5	-	-	-	-	-	-
Formaldehyde binder							
Sample 1	-	307	-	-	-	-	-
Sample 2	-	-	307	-	-	-	-
Sample 3	-	-	-	307	-	-	-
Sample 4	-	-	-	-	307	-	-
Sample 5	-	-	-	-	-	307	-
Sample 6	-	-	-	-	-	-	307
Water	<u>268</u>	<u>-</u>	<u>-</u>	<u>-</u>	<u>-</u>	<u>-</u>	<u>-</u>
Total	4000	4134	4134	4134	4134	4134	4134
Gel time in secs.	65	66	65	67	66	62	69

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and 8 secs./mm. The thickness of the boards is 17.3 mms.

5 The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 cms.

10 The results obtained are reported in the following table and are average values.

15

20

25

30

	Control	1	2	3	4	5	6
Density (kg/m ³)	688	689	687	685	690	685	692
Bending strength (N/mm ²)	19.6	19.5	18.3	19.5	20.1	19.3	19.4
Tensile strength (N/mm ²)	0.71	0.69	0.67	0.73	0.74	0.72	0.70
2 hr. thickness swelling (%)	7.0	6.5	5.0	5.5	6.1	6.5	6.2
24 hr. thickness swelling (%)	19.7	19.3	20.0	19.8	19.5	20.1	19.9
Free formaldehyde (mg/100 g dry board)	16.1	8.9	8.6	11.0	9.0	9.8	8.5
Formaldehyde reduction %	-	45	47	32	44	39	47

The above-mentioned results prove that all samples used have values equivalent to the control and that the formaldehyde reduction is of the order of 32 to 47 %.

5

Example 7

In this example three different types of organic compounds containing hydroxyl groups are exemplified by means of dextrin, phenol and resorcinol.

One monohydric alcohol acting as an additive other than methanol is also illustrated, namely ethyl alcohol.

15

The various types of formaldehyde binder used are as follows:

	1	2	3	4	
20	Monethyleneglycol	230	-	-	-
	Dextrin	-	140	-	-
	Phenol	-	-	130	-
	Resorcinol	-	-	-	130
	Methanol	-	140	130	130
25	Ethanol	80	-	-	-
	Urea	350	350	370	370
	Water	<u>340</u>	<u>370</u>	<u>370</u>	<u>370</u>
		1000	1000	1000	1000
	% solids	66	63	63	63

30

All above-mentioned figures are in parts by weight. The formaldehyde binder samples are used in this example to substitute a part of the resin used.

The glue formulations actually used are as follows:

		Control grams	1 grams	2 grams	3 grams	4 grams
5	Urea formaldehyde resin (Molar ratio F:U = 1.27:1)	3077	2770	2770	2770	2770
	Hardener (Ammonium chloride)	400	500	400	450	400
10	15 % solution)					
	Paraffin emulsion 50 %	250	250	250	250	250
	Ammonia 25° Baumé	5	-	-	-	-
	Formaldehyde binder					
	Sample 1	-	307	-	-	-
15	Sample 2	-	-	307	-	-
	Sample 3	-	-	-	307	-
	Sample 4	-	-	-	-	307
	Water	268	-	-	-	-
	Total	4000	3827	3727	3777	3727
20	Gel time in secs.	65	64	62	63	63

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and 25 8 secs./mm. The thickness of the boards is 17.3 mms. The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards pressed are 40 x 56 cms.

30 The results obtained are reported in the following table and are average values.

	Control	1	2	3	4
Density (kg/m ³)	702	698	695	705	710
Bending strength (N/mm ²)	20.1	19.5	19.7	20.0	20.8
Tensile strength (N/mm ²)	0.75	0.70	0.75	0.73	0.72
2 hr. thickness swelling (%)	7.1	6.8	6.5	6.3	6.6
24 hr. thickness swelling (%)	20.3	21.5	21.3	21.8	21.3
Free formaldehyde (mg/100 g dry board)	13.1	9.6	9.2	8.2	10
Formaldehyde reduction (%)	-	27	30	37	24

The above-mentioned results prove that all samples have values that are equivalent to the control sample that contains no formaldehyde binder even though the latter substitutes an equivalent amount of urea-formaldehyde resin in the formulation. The formaldehyde reduction varies in this example from 24 to 37 %.

Example 8

In this example one type of formaldehyde binder is used and the resin is based on phenol-melamine-urea formaldehyde resin.

The formaldehyde binder used has the following formulation:

15		<u>parts by weight</u>
	Monoethyleneglycol	300
	Urea	330
	Water	<u>370</u>
		1000

20

The glue formulations used are as follows:

		<u>Control</u>	<u>I</u>
		<u>grams</u>	<u>grams</u>
25	Phenol-melamine-urea-formaldehyde resin 63 %	5600	5600
	Hardener (solution in water of 15.5 % ammonium chloride)	840	840
	Paraffin emulsion 50 %	150	150
30	Formaldehyde binder	<u>-</u>	<u>560</u>
	Total	6590	7150
	Gel time in secs.	73	79

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kgs wood chips. Boards are pressed at 10, 9 and 8 secs./mm. The thickness of the boards is 17.3 mms.

5 The temperature of the press is 200 °C and the pressure is 35 kg/cm². The dimensions of the boards produced are 40 x 56 cms.

10 The results obtained are reported in the following table and are average values.

	Control	1
Density (kg/m ³)	705	695
Bending strength (N/mm ²)	26.2	25.9
15 Tensile strength (N/mm ²)	0.27	0.26
Tensile strength V100 (N/mm ²)	2.7	2.6
2 hr. thickness swelling (%)	7.1	6.2
24 hr. thickness swelling (%)	12.0	11.3
Free formaldehyde		
20 (mg/100 g dry board)	12.8	7
Formaldehyde reduction (%)	-	45

25 The above-mentioned results prove that the formaldehyde binder according to the present invention can be used also for phenol-melamine-urea-formaldehyde resins reducing considerably the free formaldehyde emission without adversely affecting the properties of the boards.

Example 9

In this example the formaldehyde binder used includes an inorganic compound [component (d)], illustrated in this case by sodium chloride.

5

The formaldehyde binder used has the following formulation:

		<u>Parts by weight</u>
	Monoethyleneglycol 100 %	270
10	Urea 100 %	318
	Sodium chloride 100 %	50
	Water	362
	Total	1000
15	% solids	63.8

The glue formulations used in the various samples are as follows:

	<u>Control</u>	<u>1</u>
	grams	grams
	Urea-formaldehyde resin	
	(Molar ratio F:U=1.27:1)	3077
	Hardener	2770
25	(Ammonium chloride 15 % solution)	450
	Paraffin emulsion 50 %	400
	Ammonia 25° Baumé	250
	Formaldehyde binder	5
30	Water	-
	Total	307
	Gel time in secs.	-
		3777
		64

Single-layer boards are prepared in the laboratory by spraying each of these formulations onto 25 kg wood chips. Boards are pressed at 10, 9 and 8 secs/mm. The thickness of the boards is 17.3 mm. The temperature of the press is 5 200 °C and the pressure is 35 kg/cm². The dimensions of the boards pressed are 40 x 56 cms.

The results obtained are reported in the following table and are average values.

10

	<u>Control</u>	<u>1</u>
Density (kg/m ³)	695	699
Bending strength (N/mm ²)	19.9	19.5
Tensile strength (N/mm ²)	0.71	0.73
15 2 hr. thickness swelling (%)	7.0	6.6
24 hr. thickness swelling (%)	20.5	20.8
Free formaldehyde (mg/100 g dry board)	17	9.5
Formaldehyde reduction (%)	-	44

20

The results prove that the sample including the formaldehyde binder gives values that are equivalent to the control sample in spite of the fact that the formaldehyde binder substitutes a part of the urea-formaldehyde resin in 25. the formulation. The formaldehyde reduction is 44 %.

30

C L A I M S

1) A formaldehyde binder when for use in boards prepared from lignocellulosic materials using adhesives 5 based on formaldehyde, which comprises a solution of (a) at least one organic hydroxy compound with the exception of C_{1-4} monohydric aliphatic alcohols and (b) at least one amide in water.

10

2) The formaldehyde binder of claim 1 which, in addition, contains (c) an organic compound which acts as a solvent for (a) and (b) and also reacts with formaldehyde.

15

3) The formaldehyde binder of claim 2 wherein (c) is a C_{1-4} aliphatic monohydric alcohol.

20

4) The formaldehyde binder of any one of claims 1 to 3 which, in addition, contains (d) an inorganic compound soluble in water.

25

5) The formaldehyde binder of claim 4 wherein (d) is a water-soluble halide salt.

6) The formaldehyde binder of claim 5 wherein (d) is a water-soluble halide of an alkali metal or an alkaline earth metal.

30

7) The formaldehyde binder of claim 6 wherein (d) is sodium chloride, potassium chloride or calcium chloride.

8) The formaldehyde binder of any one of claims 1 to 7 wherein component (a) is soluble in water or in a C_{1-4} monohydric aliphatic alcohol.

9) The formaldehyde binder of claim 8, wherein component (a) is selected from dihydric, trihydric and pentahydric alcohols containing up to 6 carbon atoms, monosaccharides containing up to 6 carbon atoms, di-
5 saccharides containing up to 12 carbon atoms and polysaccharides having an Ostwald viscosity up to 200 mPas at 25 °C at a concentration corresponding to 37 % (refraction).

10) The formaldehyde binder of claim 8, wherein component (a) is selected from aromatic alcohols and phenols.

11) The formaldehyde binder of claim 10, wherein component (a) is selected from monohydric and poly-
15 hydric aromatic alcohols containing one benzene ring and monohydric and polyhydric phenols.

12) The formaldehyde binder of any one of claims 1 to 11 wherein component (b) is soluble in water or in a
20 C_{1-4} monohydric aliphatic alcohol.

13) The formaldehyde binder of claim 12, wherein component (b) is selected from aliphatic amides containing up to 6 carbon atoms and aromatic amides containing
25 one benzene ring.

14) The formaldehyde binder of any one of claims 1 to 13, characterized in that the ratio by weight of component (a) plus components (c) and (d), if present, to
30 component (b) is 10:100 to 400:100.

15) The formaldehyde binder of any one of claims 1 to 14, characterized in that it contains 20 to 80 % by weight of the active ingredients.

16) The formaldehyde binder as claimed in claim
1 substantially as hereinbefore described in any of the Examples.

17) A process for preparing the formaldehyde bin-
5 der as claimed in claim 1, wherein

(a) at least one organic hydroxy compound with the
exception of C₁₋₄ monohydric aliphatic alcohols,

(b) at least one amide and

10 (c) optionally at least one organic compound which acts
as a solvent for (a) and (b) and also reacts with formalde-
hyde and

(d) optionally an inorganic compound soluble in water
and water are mixed at a temperature from room temperature
to 70°C.

15

18) A process as claimed in claim 17 wherein (c)
is a C₁₋₄ monohydric aliphatic alcohol.

19) A process as claimed in claim 17 or 18 wherein (d)
20 is a water-soluble halide salt.

20) A process as claimed in claim 19 wherein (d) is a
water-soluble halide of an alkali metal or an alkaline earth
metal.

25

21) A process as claimed in claim 20 wherein (d) is
sodium chloride, potassium chloride or calcium chloride.

22) A process according to any one of claims 17 to 21
30 wherein component (a) is soluble in water or in a
C₁₋₄ monohydric aliphatic alcohol.

23) A process according to claim 22, wherein
component (a) is selected from dihydric, trihydric and

5 pentahydric alcohols containing up to 6 carbon atoms, monosaccharides containing up to 6 carbon atoms, disaccharides containing up to 12 carbon atoms and polysaccharides having an Ostwald viscosity up to 200 mPas at 25 °C at a concentration corresponding to 37% (refraction).

24) A process according to claim 22, wherein component (a) is selected from aromatic alcohols and phenols.

10 25) A process according to claim 24, wherein component (a) is selected from monohydric and polyhydric aromatic alcohols containing one benzene ring and monohydric and polyhydric phenols.

15 26) A process according to any one of claims 17 to 25, wherein component (b) is soluble in water or in C₁-4 monohydric aliphatic alcohols.

20 27) A process according to claim 26, wherein component (b) is selected from aliphatic amides containing up to 6 carbon atoms and aromatic amides containing one benzene ring.

25 28) A process according to any one of claims 17 to 27, characterized in that the ratio by weight of component (a) plus components (c) and (d), if present, to component (b) is 10:100 to 400:100.

30 29) A process according to any one of claims 17 to 28, characterized in that the formaldehyde binder contains 20 to 80 % by weight of the active ingredients.

30) A process for preparing boards from lignocelulosic materials using an adhesive based on formaldehyde,

wherein a formaldehyde binder according to any one of claims 1 to 16 is added to the adhesive.

31) A process as claimed in claim 30 substantially 5 as hereinbefore described in the Examples.

32) Boards when produced by a process as claimed in claim 30 or claim 31.

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